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MASSACHUSETTS INST OF TECH CAMBRIDGE DEPT OF CHEMISTRY F/G 20/5  
ONE- AND TWO-PHOTON OPTICALLY PUMPED LASER DEVICES: NEW CHEMICALS--ETC(U)  
SEP 80 R W FIELD AFOSR-76-3056

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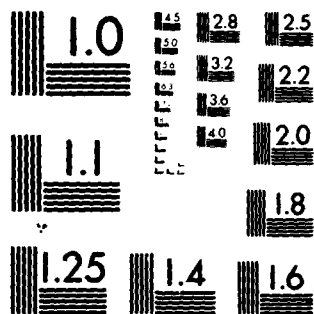
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7. AUTHOR(s) <b>Robert W. Field</b>		8. CONTRACT OR GRANT NUMBER(s) <b>AFOSR-76-3056</b>
6. PERFORMING ORGANIZATION NAME AND ADDRESS <b>Department of Chemistry Massachusetts Institute of Technology Cambridge, Massachusetts 02139</b>		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS <b>2303/B1 61102F</b>
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) <b>Research programs in four general areas have resulted in publication of 16 papers, with 10 more in preparation. These areas include: optically pumped B-X electronic transition laser; laser spectroscopy of alkaline earth halides; development of a new technique, Stimulated Emission Pumping, by which highly excited vibrational levels of combustion species may be selectively prepared and examined; optically pumped Na<sub>2</sub> B-X laser.</b>		

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## B. Research Objectives

1. Development of new optically pumped electronic transition lasers;
2. Use of optically pumped lasers to obtain spectroscopic and kinetic information in novel ways, particularly for measurement of collisional removal rates from non-fluorescing levels;
3. Investigation of the electronic structure of a family of molecules, employing novel techniques in tunable laser spectroscopy;
4. Demonstration of a new technique for selective preparation of highly excited vibrational levels of small polyatomic molecules, free of the requirements of achieving cw, laser oscillation.

## C. Status of Research Effort

All of the cited objectives have been achieved. Research in each of the areas is continuing under new AFOSR, NSF, and DoE (pending) sponsored programs.

1. The first cw, optically pumped, electronic transition laser was demonstrated. This included 1000 individual laser lines of the  $I_2$   $B^1\Omega_u-X^1\Sigma_g^+$  system, pumped by the  $Ar^+$  5145 and 5017Å lines, which span the region 5500-13500Å. In addition, various lines of our  $Ar^+$  laser pumped, cw,  $Na_2$   $B^1\Pi_u-X^1\Sigma_g^+$  laser have been made to oscillate.
2. First uses of optically pumped lasers include: observation of highly excited vibrational levels near a dissociation limit; a new and general technique for measurement of hyperfine structure in highly excited, non-fluorescing levels; a method for measurement of electronic transition moment functions based on relative gain; measurements of total collisional removal rates from non-fluorescing levels; use of an optically pumped laser as means of both preparing population in selected vibrationally excited levels and monitoring subsequent dye laser excitation from these levels to levels near dissociation limits.
3. The techniques of Optical-Optical Double Resonance, Intermodulation, and Selective Fluorescence Detected Excitation Spectroscopy have been applied to CaF, CaBr, CaI, and BaF. Relationships between spectroscopic properties have been used to determine s, p, d hybridizations, the effectiveness of various halogens in promoting such hybridization, and the electronic states have been organized into Rydberg Series. Data from these studies have been useful to groups using LIF to probe product distributions in  $M(Ca \text{ or } Ba) + X(F, Cl, Br, I)$  reactions.
4. The technique of Stimulated Emission Pumping has been demonstrated on the  $I_2$  B-X system. This is a pulsed excitation scheme whereby 30% of the population in a single rotation-vibration level may be transferred in two steps, into a single, highly excited rotation-vibration level. Any diatomic or small polyatomic molecule from which laser induced fluorescence from a single excited rovibronic level can be observed, has been shown to be subject to this excitation scheme. The structural and chemical properties of levels with 2-3eV of vibrational excitation may thus be purposefully investigated.

D. List of Publications

List of Publications Acknowledging AFOSR Support

1. R.A. Gottscho, J.B. Koffend, J.R. Lombardi, and R.W. Field, "Optical-Optical Double Resonance Laser Spectroscopy BaO. II," J. Chem. Phys. 68, 4110-4122 (1978).
2. R.A. Gottscho and R.W. Field, "Assignment of Extra Lines in a Perturbed Band Spectrum Using Power Broadened Line Widths, Chem. Phys. Lett. 60, 65-68 (1978).
3. J.B. Koffend, R. Bacis, and R.W. Field, "CW Optically Pumped Iodine Laser II. Spectroscopy and Long Range Analysis of the  $X^1\Sigma$  Ground State of  $I_2$ ", J. Mol. Spect. 77, 202-212 (1979).
4. J.B. Koffend and R.W. Field, "CW Optically Pumped Molecular Iodine Laser", J. Appl. Phys. 48, 4468-4472 (1977).
5. J.B. Koffend, F.J. Wodarczyk, and R.W. Field, pages 96-109, High Power Lasers and Applications, K.L. Kompa and H. Walther (editors), Springer Verlag (1978).
6. J.B. Koffend, R.W. Field, D.R. Guyer, and S.R. Leone, "Pulsed and CW Optically Pumped Lasers for Novel Applications in Spectroscopy and Kinetics", pages 382-393, Laser Spectroscopy III, J.L. Hall and J.L. Carlsten (editors), Springer Verlag, 1977.
7. R.W. Field, "Transitions Near Dissociation Limits by Optical Optical Double Resonance and Optically Pumped Laser Spectroscopy", pages 143-155, États Atomiques et Moléculaires Couplés a un Continuum. Atomes et Molécules Hautement Excités, Editions du CNRS No. 273 (1977).
8. J.B. Koffend, S. Goldstein, R. Bacis, R.W. Field, and S. Ezekiel, "Doppler-Free Emission Spectroscopy and Secondary Frequency Standards Using an Optically Pumped Laser," Phys. Rev. Lett. 41, 1040-1045 (1978).
9. J. Brooke Koffend, R. Bacis, and Robert W. Field, "The Electronic Transition Moment of the  $BO^+-X^1\Sigma^+$  System of  $I_2$  Through Gain Measurements of an  $I_2$  Optically Pumped Laser", J. Chem. Phys. 70, 2366-2372 (1979).
10. J. Brooke Koffend, Roger Bacis, and Robert W. Field, "Stimulated Emission Spectroscopy with an Optically Pumped  $I_2$  Laser", SPIE Proceedings for the International Conference on Lasers '78, Volume 168, pages 000-000, 1979.
11. J.B. Koffend, F.J. Wodarczyk, R. Bacis, and R.W. Field, "Collisional Relaxation of Highly Excited Vibrational Levels of the  $I_2 X^1\Sigma^g$  State Using an  $I_2$  Optically Pumped Laser", J. Chem. Phys. 72, 478-483 (1980).

Availability Codes		
Dist.	Avail and/or special	
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12. P.F. Bernath and R.W. Field, "Optical-Optical Double Resonance Spectroscopy of CaF: The  $E^2\Sigma^+$  and  $E'^2\Pi$  States", J. Mol. Spectry. 00, 000-000 (1980).
13. R. Bacis, S. Churassy, R.W. Field, J.B. Koffend, and J. Verges, "High Resolution and Sub-Doppler Fourier Transform Spectroscopy: Iodine Molecular Fluorescence Excited by the 514.5 and 501.7 nm  $Ar^+$  Laser Lines", J. Chem. Phys. 72, 34-42 (1980).
14. M. Dulick, P.F. Bernath, and R.W. Field, "Rotational and Vibrational Analysis of the CaF  $B^2\Sigma^+-X^2\Sigma^+$  System" Can. J. Phys. 58, 703-712 (1980).
15. P.F. Bernath, M. Dulick, R.W. Field, and J. Hardwick, "Laser Excited Fluorescence of  $CS_2$ " J. Mol. Spectrosc. 00, 000-000 (1980).
16. P.F. Bernath, P.G. Cummins, and R.W. Field, "Intermodulated Fluorescence Spectroscopy of CaF  $A^2\Pi-X^2\Sigma^+$ ", Chem. Phys. Lett. 70, 618-620 (1980).

List of AFOSR-Supported Papers in Preparation

17. P. F. Bernath, R. W. Field, B. Pinchemel, Y. Lefebvre, and J. Schamps, "Laser Spectroscopy of CaBr:  $A^2\Pi-X^2\Sigma^+$  and  $B^2\Sigma^+-X^2\Sigma^+$  Transitions", for submission to J. Mol. Spectrosc.
18. P. F. Bernath, B. Pinchemel, and R. W. Field, "The Hyperfine Structure of the Calcium Halides", for submission to J. Chem. Phys.
19. P. F. Bernath, B. Pinchemel, R. W. Field, K. Möller, and T. Törring, "Simultaneous Fitting of Optical and Microwave Data: CaBr as Example", for submission to J. Mol. Spectrosc.
20. P. Ip, P. F. Bernath, and R. W. Field, "Optical-Optical Double Resonance Spectroscopy of BaF: The  $E^2\Sigma^+$  and  $F^2\Pi$  States", for submission to J. Mol. Spectrosc.
21. D. Reisner, P. F. Bernath, and R. W. Field, "Laser Spectroscopy of CaI:  $A^2\Pi-X^2\Sigma^+$  and  $B^2\Sigma^+-X^2\Sigma^+$  Transitions", for submission to J. Mol. Spectrosc.
22. P. F. Bernath and R.W. Field, "The Electronic Structure of the Calcium Halides", for submission to J. Chem. Phys.
23. R. F. Marks and R. W. Field, "The CaO  $a^3\Pi$  and  $A'^1\Pi$  States from Partial Analysis of the 'Orange' Band System", for submission to J. Chem. Phys.
24. R. Bacis, S. Churassy, J. Vergés, and R. W. Field, "The  $I_2$   $0_g^+$  and  $1_g$  States by LIF-FTS", for submission to J. Chem. Phys.
25. R. Bacis, D. Cerny, R. W. Field, and R. A. McFarlane, "Assignment and Kinetic Mechanism for  $N_2$   $W^3\Delta-B^3\Pi$  and  $B^3\Pi-W^3\Delta$  Lasing Lines", for submission to J. Mol Spectrosc.
26. C. Kittrell, E. Abramson, D. H. Katayama, S. McDonald, D. E. Reisner, and R. W. Field, "Stimulated Emission Pumping of  $I_2$   $BO_u^+-X^1\Sigma_g^+$ ", for submission to J. Chem. Phys.

E. Personnel

1. Visiting Scientists

Dr. Roger Bacis (optically pumped  $I_2$  laser)  
Université Claude Bernard (Lyon I)  
Villeurbanne, FRANCE

Dr. Francis J. Wodarczyk (optically pumped  $I_2$  laser)  
Rome Air Development Center (ESO)  
Hanscom Air Force Base

Dr. Daniel Katayama (stimulated emission pumping)  
Air Force Geophysics Laboratory  
Hanscom Air Force Base

2. Postdocs

Dr. Philip G. Cummins (alkaline earth halides)  
Dr. Ingemar Renhorn (alkaline earth halides)  
Dr. Hartmut Schweda ( $Na_2$  laser)  
Dr. Carter Kittrell (stimulated emission pumping)

3. Graduate Students

Dr. J. Brooke Koffend\* (optically pumped  $I_2$  laser)  
Ph.D. November 1978

Dr. Peter F. Bernath\* (alkaline earth halides)  
Ph.D. November 1980

Ms. Gunjit Chawla ( $Na_2$  laser)

Dr. Michael Dulick (alkaline earth halides)  
Ph.D. February 1981

Dr. Richard A. Gottscho (alkaline earth halides)  
Ph.D. February 1979

Ms. Precila Ip (alkaline earth halides)

Mr. Ronald Marks (computer controlled laser spectroscopy)

Mr. Bernard Pinchemel (alkaline earth halides)  
Visitor from Université des Sciences et Techniques de Lille, FRANCE  
Ph.D. expected June 1981

Mr. Evan Abramson (stimulated emission pumping)

Mr. Steve McDonald (stimulated emission pumping)

Mr. David E. Reisner (stimulated emission pumping)

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\* Major contributor to AFOSR research programs.

4. Undergraduates

Mr. Philip Brucat  
B.S. May 1977

Mr. Alain Drooz  
B.S. May 1980

Mr. Jeffrey Manni  
B.S. May 1978

Mr. Hrvoje Petek  
B.S. May 1980

Mr. Andrew Ralston

Mr. Paul S. Weiss  
B.S. and M.S. May 1980

Mr. Michael Wilson

F. Interactions: Spoken Papers

6-77 Molecular Spectroscopy Symposium, Columbus, Ohio, papers WF5, WF10.

P.G. Cummins: "Laser Induced Fluorescence Spectrum of BaF".

J.B. Koffend: "CW Optically Pumped I<sub>2</sub> Laser".

6-77 Observatoire de Meudon, France.

Invited Talk, "Stepwise Excitation of Small Polyatomic Molecules to Energy Levels Close to a Dissociation Limit".

6-77 Aussois, France, Centre National Recherche Scientifique Colloque International #273.

Invited Talk, "Transitions Near Dissociation Limits by OODR and OPL Spectroscopy".

6-77 Fourth Colloquium on Electronic Transition Lasers, Munich, Germany.

Invited Talk "Optically Pumped Molecular I<sub>2</sub> Laser".

10-77 M.I.T. Electrical Engineering and Computer Science Seminar Series.

"A CW Optically Pumped I<sub>2</sub> Laser"

11-77 University of Maryland, Institute for Physical Science and Technology.

"Electronic Transition Optically Pumped Lasers".



- 1-78 Allied Chemical, Photon Chemistry Group.  
"Electronic Transition Optically Pumped Lasers"
- 2-78 Dalhousie University, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy with Diatomic Molecules".
- 2-78 University of New Brunswick, Physics Department.  
"Electronic Transition Optically Pumped Lasers".
- 3-78 M.I.T. Optics and Spectroscopy Seminar Series.  
"Diatomic Laser Spectroscopy. Sharply Focused and Without Ambiguity".
- 5-78 University of Illinois, Department of Chemistry.  
"Optically Pumped Lasers".
- 6-78 Molecular Spectroscopy Symposium, Columbus, Ohio, papers MF2, MF3, RF8.  
  
R. Bacis: "High Resolution Fourier Spectroscopy of the  $\text{Ar}^+$  Laser 5145Å Excited  $\text{I}_2$  Fluorescence Spectrum".  
  
J.B. Koffend: "Stimulated Emission Spectroscopy of  $\text{I}_2$ : Electronic Transition Moment of the B-X System Between 2.4-4.2Å and Hyperfine Structure of  $X^1\Sigma_g^+ v'' = 83$ ".  
  
P.F. Bernath: "Experimental and Theoretical Investigation of the Electronic Structure of  $\text{CaF}$ ".
- 7-78 DuPont Central Research and Development.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 9-78 Wesleyan University, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 11-78 Boston College, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 12-78 Lasers '78, Orlando, Florida.  
  
J.B. Koffend: "Stimulated Emission Spectroscopy with an Optically Pumped  $\text{I}_2$  Laser".

- 1-79 University of Toronto, Department of Physics.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 2-79 Harvard University, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy"
- 2-79 State University of New York at Binghamton, Department of Chemistry.  
"Double Resonance, Stimulated Emission, and Fourier Transform Spectroscopy".
- 3-79 Cornell University, Department of Chemistry  
"Double Resonance and Stimulated Emission Spectroscopy".
- 3-79 University of Pennsylvania, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 3-79 Pennsylvania State University, Department of Chemistry.  
"Stimulated Emission and Fourier Transform Spectroscopy: Iodine Re-revisited".
- 3-79 National Research Council of Canada, Herzberg Institute for Astrophysics.  
"More than You Can Say in One Hour on OODR, OPL and Related Matters".
- 3-79 McMaster University, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy with Diatomic Molecules".
- 3-79 University of Waterloo, Physical Chemistry.  
"Stimulated Emission and Fourier Transform Spectroscopy: Iodine Re-revisited".
- 4-79 Brown University, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 4-79 Université de Sherbrooke, Department of Chemistry.  
"Double Resonance and Stimulated Emission Spectroscopy with Diatomic Molecules".
- 4-79 Air Force Geophysics Laboratory, Hanscom AFB.  
"What You Wanted to Know About Spectroscopic Properties of Small Molecules but Were Afraid to Ask".

- 6-79 Canadian Association of Physicists, Vancouver, B.C.  
Plenary Lecture, "Double Resonance, Saturation, and Stimulated Emission Spectroscopy".
- 7-79 EUCHEM Conference on Molecular Electronic Spectroscopy, Cirencester, ENGLAND.  
Plenary Lecture, "Sub-Doppler Spectroscopy".
- 7-79 Université des Sciences et Techniques de Lille, FRANCE.  
"Double Resonance and Stimulated Emission Spectroscopy".
- 8-79 Université Claude Bernard, Lyon, FRANCE.  
"Doppler-Free Spectroscopy".
- 9-79 University of Tokyo (Komaba), Department of Pure and Applied Sciences.  
"Fourier Transform-Laser Induced Fluorescence Spectroscopy".
- 9-79 Sixth Colloquium on High Resolution Molecular Spectroscopy, Tours, FRANCE. Talk J15.  
B. Pouilly, B. Pinchemel, J. Schamps, P.F. Bernath, and R.W. Field,  
"Electronic Spectroscopy of CaF".
- 10-79 AFOSR/FJSRL Molecular Dynamics Conference, UASF Academy.  
"From Optically Pumped Lasers to Stimulated Emission Pumping".
- 10-79 Tri-Joint Chemical Physics Seminar (UCLA, USC, Cal. Tech.).  
Los Angeles, California.  
"Stimulated Emission Spectroscopy".
- 10-79 Stanford University, Department of Chemistry, Stanford, California.  
"Stimulated Emission Spectroscopy"
- 1-80 Brookhaven National Laboratory, Department of Chemistry.  
"Stimulated Emission Spectroscopy".
- 2-80 Amherst College, Department of Chemistry.  
"Stimulated Emission Spectroscopy of Small Molecules".
- 3-80 Memorial University of Newfoundland, Department of Physics.  
"From Optically Pumped Lasers to Stimulated Emission Pumping".

- 3-80 American Physical Society, H.P. Broida Award in Atomic and Molecular Physics, Talk AA1.

"From Optically Pumped Lasers to Stimulated Emission Pumping".

- 6-80 Molecular Spectroscopy Symposium, Columbus, Ohio. Papers MF7, MS5, MF6, TG12, WG4.

P.F. Bernath and R.W. Field, "Electronic Structure of the Alkaline Earth Halides".

P.F. Bernath, P. Ip, and R.W. Field, "Optical-Optical Double Resonance Spectroscopy of BaF".

D.E. Reisner, P.F. Bernath, and R.W. Field, "Rotational Analysis of the  $B^2\Sigma^+ - X^2\Sigma^+$  Band System of CaI".

S. Churassy, R. Bacis, R.W. Field, F. Martin, and J. Verges, "Deperturbation of Two Weakly Bound States of the Iodine Molecule:  $1_g$  and  $0_g^+$ ".

C. Kittrell, D.H. Katayama, and R.W. Field, "Stimulated Emission Pumping (SEP) of  $I_2$   $B0_u^+ - X^1\Sigma_g$  with Pulsed Dye Lasers".

- 7-80 United Technologies Research Center, Chemical Physics.

"From Optically Pumped Lasers to Stimulated Emission Pumping".

- 7-80 Exxon Corporate Research Laboratories.

"From Optically Pumped Lasers to Stimulated Emission Pumping".

- 7-80 Gordon Conference on Atomic and Molecular Interactions.

"Spectroscopic Determination of the Long-Range Behavior of the  $X^1\Sigma_g^+$ ,  $0_g^+$ , and  $1_g$  States of  $I_2$ ".

#### G. Patents

One patent applied for, covering laser pumped, electronic transition, cw, dimer lasers.

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